Supporting Information for: "Variational formulation of the generalized many-body expansion with self-consistent charge embedding: Simple and correct analytic energy gradient for fragment-based *ab initio* molecular dynamics"

Jie Liu,* Bhaskar Rana, Kuan-Yu Liu, and John M. Herbert†
Department of Chemistry and Biochemistry, The Ohio State University, Columbus, OH 43210
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TABLE S1: Difference (in hartrees/bohr) between analytic and finite-difference gradients for the variational EE-GMBE(1) method, computed at the B3LYP/STO-3G level for $(H_2O)_{10}$. Fragmentation was performed using $\xi = 3.0$ Å, resulting in 12 fragments with 6 water molecules per fragment.

Atom	x	y	z
O	−0.000001	0.000006	−0.000001
H	0.000000	0.000000	0.000000
H	0.000004	0.000012	0.000002
O	0.000001	−0.000003	0.000002
H	0.000001	−0.000011	0.000007
H	0.000000	0.000001	−0.000001
O	0.000000	−0.000006	−0.000002
H	0.000000	0.000001	0.000000
H	0.000004	−0.000008	−0.000005
O	−0.000002	−0.000005	0.000003
H	0.000001	0.000000	0.000000
H	0.000000	0.000000	−0.000001
O	0.000002	0.000002	−0.000006
H	0.000000	0.000003	−0.000011
H	0.000000	−0.000001	0.000001
O	−0.000001	−0.000003	−0.000002
H	0.000000	0.000001	0.000000
H	0.000001	0.000000	0.000000
O	0.000003	0.000001	−0.000003
H	0.000000	0.000000	0.000000
H	−0.000001	0.000000	0.000000
O	0.000000	0.000000	0.000002
H	0.000000	−0.000001	0.000000
H	−0.000006	0.000002	0.000007
O	−0.000003	0.000005	0.000001
H	0.000000	0.000000	0.000000
H	0.000001	−0.000001	0.000000
O	−0.000001	0.000004	0.000006
H	−0.000001	0.000000	0.000000
H	0.000000	−0.000001	0.000000
MAD[a]	0.000001	0.000003	0.000002

[a] Mean absolute deviation (from zero), averaged over all atoms.

* Present address: Hefei National Laboratory of Physical Science at the Microscale, University of Science and Technology of China, Anhui 230026, China
† herbert@chemistry.ohio-state.edu

TABLE S2: Difference (in hartrees/bohr) between analytic and finite-difference gradients for the variational EE-GMBE(1) method, computed at the B3LYP/STO-3G level for $F^-(H_2O)_{10}$.

Atom	x	y	z
O	0.000009	0.000001	0.000006
H	0.000001	0.000001	0.000002
H	0.000000	0.000000	0.000000
O	0.000006	−0.000010	0.000017
H	−0.000004	0.000003	−0.000002
H	0.000000	−0.000003	−0.000001
O	0.000004	−0.000014	0.000004
H	−0.000001	0.000001	0.000001
H	−0.000001	−0.000005	0.000001
O	0.000009	0.000008	0.000002
H	0.000000	0.000004	0.000001
H	0.000001	0.000000	−0.000001
O	0.000004	0.000015	0.000000
H	0.000001	0.000004	0.000002
H	−0.000002	0.000001	0.000000
O	−0.000003	−0.000007	−0.000026
H	−0.000001	0.000001	−0.000002
H	−0.000001	−0.000003	−0.000005
O	−0.000007	−0.000008	−0.000010
H	−0.000001	−0.000007	−0.000003
H	−0.000001	−0.000001	−0.000002
O	−0.000004	0.000012	0.000001
H	0.000001	−0.000003	0.000000
H	−0.000011	0.000003	−0.000005
O	−0.000001	0.000000	−0.000002
H	0.000001	0.000001	−0.000001
H	0.000000	0.000002	−0.000002
O	0.000001	0.000003	0.000011
H	−0.000001	0.000001	0.000009
H	0.000001	0.000000	0.000004
F	−0.000003	−0.000001	0.000002
MAD[a]	0.000003	0.000004	0.000004

[a] Mean absolute deviation (from zero), averaged over all atoms.

TABLE S3: Comparison of Mulliken charges for $(H_2O)_{10}$ derived from the variational EE-GMBE density (ρ_{GMBE} in eq. 16) and the supersystem density, both computed at the B3LYP/6-31G* level of theory. The structure is the one shown in Fig. 2. Fragmentation was performed using $\xi = 3.0$ Å, resulting in 12 fragments with 6 water molecules per fragment.

Atom	Mulliken Charge		difference
	Supersystem	GMBE	
O	−0.822863	−0.826387	0.0035
H	0.436593	0.444150	−0.0076
H	0.407352	0.406144	0.0012
O	−0.815646	−0.817651	0.0020
H	0.410283	0.408977	0.0013
H	0.447633	0.452119	−0.0045
O	−0.824801	−0.826451	0.0017
H	0.440874	0.444718	−0.0038
H	0.411164	0.409104	0.0021
O	−0.876661	−0.887129	0.0105
H	0.416686	0.416337	0.0003
H	0.429259	0.435281	−0.0060
O	−0.822963	−0.826199	0.0032
H	0.406631	0.405302	0.0013
H	0.437005	0.443870	−0.0069
O	−0.871915	−0.877409	0.0055
H	0.424690	0.428664	−0.0040
H	0.408326	0.409854	−0.0015
O	−0.875977	−0.882135	0.0062
H	0.432015	0.436520	−0.0045
H	0.425082	0.424045	0.0010
O	−0.824384	−0.826262	0.0019
H	0.435800	0.444345	−0.0085
H	0.406805	0.407852	−0.0010
O	−0.874308	−0.879910	0.0056
H	0.431072	0.435638	−0.0046
H	0.420776	0.419501	0.0013
O	−0.873616	−0.878648	0.0050
H	0.424054	0.422581	0.0015
H	0.431033	0.433181	−0.0021
MAD[a]			0.0037

[a] Mean absolute deviation over all atoms.

TABLE S4: Comparison of Mulliken charges for $F^-(H_2O)_{10}$ derived from the variational EE-GMBE density (ρ_{GMBE}, eq. 16) and the supersystem density, both computed at the B3LYP/6-31G* level of theory.

Atom	Mulliken Charge		difference
	Supersystem	GMBE	
O	−0.855062	−0.853878	−0.0012
H	0.383461	0.383079	0.0004
H	0.410594	0.411490	−0.0009
O	−0.814255	−0.817740	0.0035
H	0.380763	0.387705	−0.0069
H	0.419823	0.420861	−0.0010
O	−0.828944	−0.826072	−0.0029
H	0.393625	0.394602	−0.0010
H	0.386411	0.387369	−0.0010
O	−0.824010	−0.827445	0.0034
H	0.384243	0.383547	0.0007
H	0.430617	0.433779	−0.0032
O	−0.888530	−0.887449	−0.0011
H	0.396780	0.392240	0.0045
H	0.415587	0.413483	0.0021
O	−0.842989	−0.845428	0.0024
H	0.398211	0.398887	−0.0007
H	0.392089	0.392144	−0.0001
O	−0.833188	−0.834038	0.0008
H	0.372296	0.373079	−0.0008
H	0.403425	0.404134	−0.0007
O	−0.789926	−0.789143	−0.0008
H	0.393489	0.394577	−0.0011
H	0.373222	0.374491	−0.0013
O	−0.857562	−0.857544	−0.0000
H	0.386246	0.380864	0.0054
H	0.396764	0.397577	−0.0008
O	−0.855341	−0.855672	0.0003
H	0.400629	0.400518	0.0001
H	0.395550	0.400128	−0.0046
F	−0.524018	−0.530144	0.0061
MAD[a]			0.0020

[a] Mean absolute deviation, averaged over all atoms.